APPENDIX A

SUPPLEMENTARY INFORMATION

SUPPLEMENTARY INFORMATION

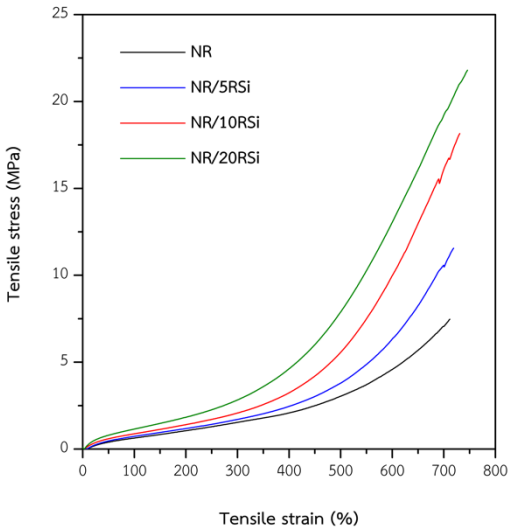

Figure A.1 Stress-strain curves of NR and NR/RSi composites.

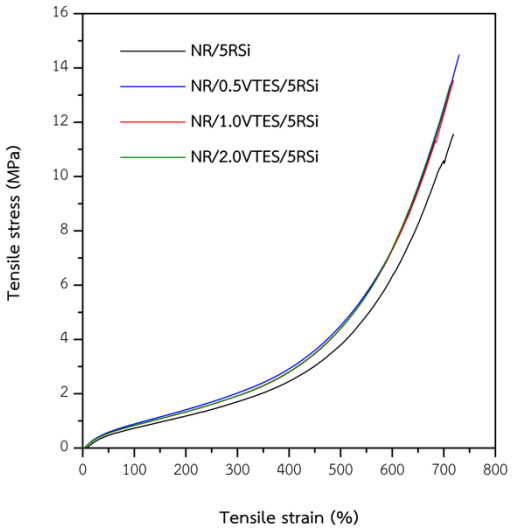

Figure A.2 Stress-strain curves of NR/5RSi and NR/VTES/5RSi composites.

SUPPLEMENTARY INFORMATION (Continued)

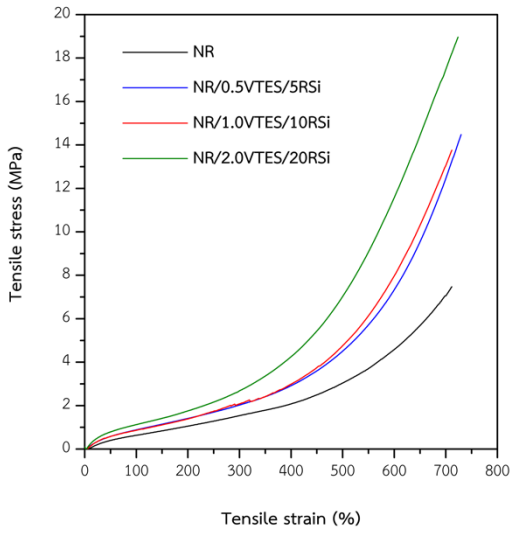

Figure A.3 Stress-strain curves of NR and NR/VTES/RSi composites.

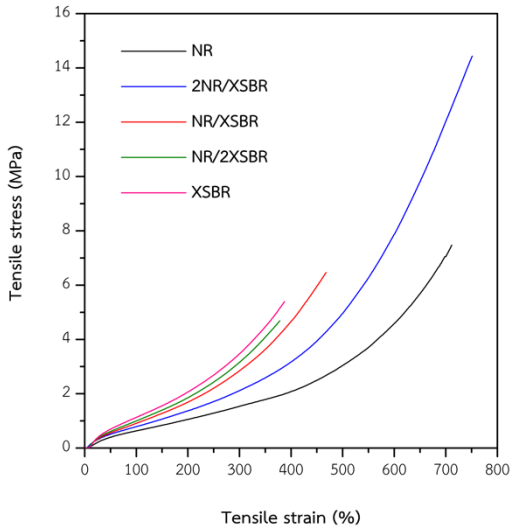

Figure A.4 Stress-strain curves of NR, XSBR, and their blends.

SUPPLEMENTARY INFORMATION (Continued)

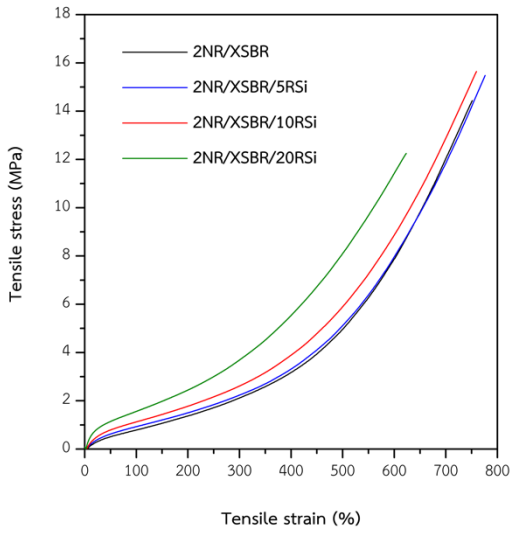

Figure A.5 Stress-strain curves of 2NR/XSBR and 2NR/XSBR/RSi composites.

Table A.1 Atomic percentages of unadded PMM and PMM.

Sample	Element (%)					
	C	O	Mg	Al	Si	Ca
Unadded PMM	15.71	64.83	0.24	1.04	3.72	14.47
0.05PMM	22.94	58.15	0.24	0.96	4.87	12.85
0.10PMM	23.87	54.16	0.66	1.20	6.31	13.81
0.20PMM	30.79	53.56	0.27	1.01	3.97	10.39

APPENDIX B

PUBLICATIONS

PUBLICATIONS

Bureewong, N., Ruksakulpiwat, Y., & Ruksakulpiwat, C. (2022a). IN SITU SILICA REINFORCED RUBBER LATEX COMPOSITE: EFFECTS OF BLEND RATIO AND SILICA CONTENT. *Suranaree Journal of Science & Technology, 29*(2).

Bureewong, N., Ruksakulpiwat, Y., & Ruksakulpiwat, C. (2022b). *Mechanical and thermal properties of NR/XSBR composite reinforced with rice husk silica.* Paper presented at the Journal of Physics: Conference Series.

IN SITU SILICA REINFORCED RUBBER LATEX COMPOSITE: EFFECTS OF BLEND RATIO AND SILICA CONTENT

Namthip Bureewong[1,2], Yupaporn Ruksakulpiwat[1,2] and Chaiwat Ruksakulpiwat[1,2,*]

Received: February 01, 2021; Revised: April 04, 2021; Accepted: April 04, 2021

Abstract

The effects of blend ratio and silica content were studied on mechanical, morphological, and thermal properties of rubber blends and composites. Natural rubber (NR) was blended with carboxylated styrene-butadiene rubber (XSBR) at different ratios of NR/XSBR, i.e., 1:1, 1:2, and 2:1. The optimum mechanical properties of NR/XSBR blends was obtained from NR/XSBR at the optimum ratio of 2:1. NR/XBSR with the optimum ratio was mixed with silica in silicate form. Commercial sodium silicate (CSS) was used to prepare the silica for use as reinforcing filler. NR/XSBR was firstly mixed with CSS to obtain the mixture of NR/XSBR/CSS. And then, acetic acid was dropped into the mixture to obtain the precipitated silica at various contents of silica (5, 10, and 20 phr). The addition of silica into NR/XSBR at the ratio of 2:1 can improve the thermal property of the NR/XSBR blends. By increasing silica contents, an insignificant difference in terms of tensile strength and elongation at break of the mechanical properties was observed. The best thermal properties were obtained from NR/XSBR/20Si composite.

Keywords: Natural rubber latex, Carboxylated styrene-butadiene rubber latex, Rubber blend, Silica, Rubber composite

Introduction

Natural rubber (NR) is one of the important agricultural products of Thailand that can be used to produce a wide of products due to high resilience, good tensile strength, and tear resistance, etc. (Chuayjuljit *et al*., 2015; Vu *et al*., 2015). The making of NR products covered many industries such as automotive (tires, belts, and seals), medical (gloves, tubes, and condoms), and others (shoes, furniture, and sponges), etc. (Rajan *et al*., 2006). However, NR is known that has low resistance for sunlight, ozone, and oxygen or heat aging because it contains the double bonds in the molecular structure (Jones and Tinker, 1997; Vu *et al*., 2015). Therefore, the most productive of NR production are not preferred to use only NR but will be used with other rubbers or fillers to obtain the properties of the product required (Varkey *et al*., 2000; Motaung *et al*., 2011). The samples of manufacturing of tires

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were applied for NR with butadiene rubber (BR) or styrene-butadiene rubber (SBR), carbon black, and silica for improving wear, traction, and rolling resistance, etc. (Le *et al*., 2010; Shan *et al*., 2011).

The other rubbers and fillers, SBR and silica are interesting because they can improve some properties of NR such as mechanical and thermal properties. Varkey *et al*. (2000) studied the thermal degradation of NR/SBR latex blends by using the thermogravimetric method and they found that blending with SBR increased the thermal stability of NR. Poompradub *et al*. (2014) reported that the filling NR with in situ silica can not improve only the mechanical properties but also the thermal properties of the filled vulcanizates, and was superior compared with the ex-situ silica. SBR is the largest volume of synthetic rubber. It can polymerize by using the solution or emulsion technique. Moreover, it has the low reversion of resistance, high resistance of abrasion at higher speeds, and good resistance for flexible fatigue. However, the main drawbacks of SBR are poor oil and ozone resistances. The carboxylated SBR (XSBR) is one of the members of SBR polymerized with the emulsion polymerization of styrene, butadiene, and some strong acid monomer that like acrylic acid or methyl methacrylate acid. It has the good abrasion resistance, aging stability, hydrocarbon solvent resistance, and interaction with the functional filler that like silica (Alimardani and Abbassi-sourki, 2014). There are several reports that mentioned about the NR blended with XSBR to easily modify the mechanical properties of NR because of the polarity of XSBR (Stephen *et al*., 2003). Also, the incorporation of XSBR into NR improves the thermal properties and gas permeability of NR (Stephen *et al*., 2005). Silica is the important filler in the rubber industry that reduces heat buildup and improves the tensile strength, tear strength, and abrasion resistance of rubber composites (Ikeda and Kohjiya, 1997; Kohjiya and Ikeda, 2003). Moreover, it is known that in the tire industry, silica is extensively used to substitute with carbon black for reducing the rolling resistance of tire treads and enhance wet grip (Alimardani and Abbassi-sourki, 2014). Nevertheless, the above reports generally mentioned the mixing of rubber in a latex form which compounding of latex is the low energy of processing of consumption but normal interaction between filler and rubber that not good because there is not present the mastication step (Stephen *et al*., 2006). Therefore, using of XSBR blended with NR composites may be solved this problem because it contained carboxyl groups which may be reduced the mastication step and compatible with silica that used as the filler in this study.

The aim of this research is focused on study of the effects of blend ratio between NR and XSBR at the ratio of 1:1, 1:2, and 2:1 on the mechanical, morphological, and thermal properties. The optimum NR/XSBR ratio was mixed with in situ silica at various contents (5, 10, and 20 phr). Mechanical, morphological, and thermal properties of NR/XSBR/Si composites were studied.

Materials and Methods

Materials

Natural rubber latex (NRL) with a solids content of 60wt% and high ammonia-treated latex was supplied by Chemical & Materials Co., Ltd. Carboxylated styrene-butadiene rubber latex (XSBRL) was supplied by Jorakay Corporation Co., Ltd. Commercial sodium silicate (CSS) was supplied by Sigma-Aldrich Co., Ltd. Acetic acid was supplied by Merck & Co., Inc. Zinc oxide (ZnO), stearic acid, N-Cyclohexyl-2-Benzothiazole Sulfonamide (CBS), and sulfur were supported by Chemical Innovation Co., Ltd.

Preparation of NR/XSBR Sheets

NRL was blended with XSBRL at different ratios of NR/XSBR, i.e., 1:1, 1:2, and 2:1 by using a magnetic stirrer for 30 mins. Rubber mixtures were poured onto the Teflon tray and dried at 60°C by using a hot air oven for 16 hr to obtain NR/XSBR sheets.

Preparation of NR/XSBR/Si Sheets

NR/XSBR at the ratio of 2:1 was mixed with CSS at various contents of silica (5, 10 and 20 phr) by using a magnetic stirrer for 30 mins to obtain NR/XSBR/CSS mixtures. Acetic acid was dropped into the NR/XSBR/CSS mixture until neutral to gain precipitated silica. The mixtures were poured onto the Teflon tray and dried at 60°C by using a hot air oven for 16 h to obtain NR/XSBR/Si sheets.

Preparation of NR/XSBR Blends and NR/XSBR/Si Composites

All samples were mixed with 5 phr ZnO, 2 phr stearic acid, 2.5 phr CBS, and 1 phr sulfur by using a two-roll mill machine to obtain compounds. Vulcanization of all compounds was done by using the compression molding machine at 150°C with optimum curing time (t_c90) that determined by Moving Die Rheometer (MDR) to obtain NR/XSBR blends and NR/XSBR/Si composites.

Mechanical Properties

The tensile properties of blends and composites were measured according to ASTM

D412 using a Universal Testing Machine (Instron 5565) with a crosshead speed of 500 mm/min and a load cell of 5 kN.

The hardness of blends and composites were measured according to ASTM D2240 using a Bareiss shore A (HPE II).

Morphological Properties

The morphology of blends and composites were investigated by Field Emission Scanning Electron Microscope (FE-SEM) using a Carl Zeiss (Auriga). All of the samples were coated with gold before testing.

Thermal Properties

The thermal stability of blends and composites were determined by thermogravimetric analysis (TGA) using a Mettler Toledo (TGA/DSC1). All samples were placed into an alumina pan and heated at the room temperature up to 800°C under nitrogen at a heating rate of 10°C /min.

Results and Discussion

Mechanical Properties of NR/XSBR Blends

The mechanical properties of NR, XSBR, and NR/XSBR at different ratios of blending are shown in Figure 1. All NR/XSBR blends showed higher modulus at 100% and 300% elongation than NR but lower than XSBR depending on the ratios of blending. This is because XSBR has a styrene content for making the XSBR harder and rubbery less than NR. NR/XSBR at the ratio of 2:1 showed the highest tensile strength and elongation at break due to its strain-induced crystallization (Stephen *et al*., 2003). However, all NR/XSBR blends showed an insignificant difference of hardness.

Morphological Properties of NR/XSBR Blends

The morphological properties of NR, XSBR, and NR/XSBR at different ratios of blending are shown in Figure 2. The surface fracture by using the tensile testing of XSBR showed the flattest surface when compared with NR and NR/XSBR at different ratios of blending. The surface roughness of NR/XSBR increased with NR content. The highest surface roughness was obtained from NR/XSBR at the ratio of 2:1. The rough fractured surface indicate that the samples was failed in a ductile manner which led to the good elongation at break (Boonmahitthisud *et al*., 2017).

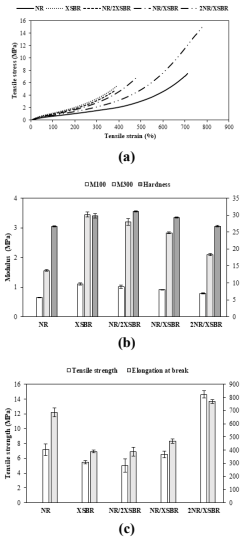

Figure 1. Mechanical properties of NR, XSBR, and NR/XSBR blends showing (a) stress-strain curve, (b) modulus at 100% elongation, modulus at 300% elongation, and hardness, and (c) tensile strength and elongation at break.

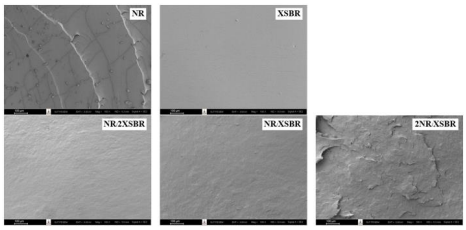

Figure 2. SEM images of NR, XSBR, and NR/XSBR blends.

Table 1. Degradation temperature at different weight loss levels of NR, XSBR, NR/XSBR blends, NR/XSBR/Si composites, and silica.

Sample	$T_{30\%}$ (°C)	$T_{50\%}$ (°C)	$T_{70\%}$ (°C)	T_{max} (°C)
NR/XSBR Blends				
NR	366	380	397	378
XSBR	401	421	442	415
2NR/XSBR	371	390	415	388
NR/XSBR/Si Composites				
2NR/XSBR/5Si	373	392	421	389
2NR/XSBR/10Si	376	400	433	390
2NR/XSBR/20Si	371	397	434	389
Silica	449	N/A	N/A	483

Thermal Properties of NR/XSBR Blends

The thermal properties in term of degradation of temperature at different levels of weight loss of NR, XSBR, and NR/XSBR at the ratios of 2:1 are listed in Table 1. NR showed the lowest degradation temperature compared with XSBR and NR/XSBR at the ratio of 2:1 because NR is a diene rubber that is highly susceptibility to degrade (Stephen *et al*., 2006). NR/XSBR at the ratio of 2:1 showed higher degradation temperature than NR while XSBR showed the highest degradation temperature. XSBR can increase the degradation temperature of the NR/XSBR blend.

Mechanical Properties of NR/ XSBR/ Si Composites

NR/XSBR at the ratio of 2:1 was selected to be blended with silica at the different contents.

The mechanical properties of NR/XSBR and NR/XSBR blended with silica at different contents are shown in Figure 3. The modulus at 100% and 300% elongation of all NR/XSBR blended with silica were higher than those of NR/XSBR blends. With increasing silica contents, modulus at 100% and 300% elongation were higher due to the higher stiffness of fillers. That is the reason of the results of the hardness of NR/XSBR and NR/XSBR blended with silica at the different contents as well. However, NR/XSBR and NR/XSBR blended with silica at different contents showed a slight difference in tensile strength and elongation at break.

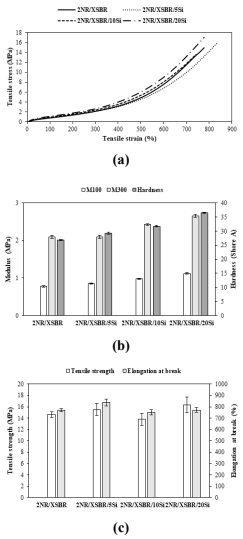

Figure 3. Mechanical properties of NR/XSBR blend and NR/XSBR/Si composites showing (a) stress-strain curve, (b) modulus at 100% elongation, modulus at 300% elongation, and hardness, and (c) tensile strength and elongation at break.

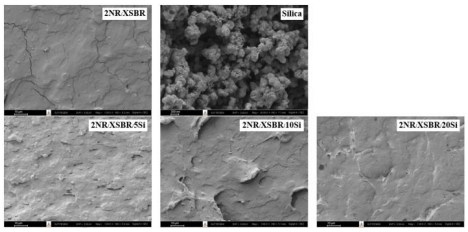

Figure 4. SEM images of NR/XSBR blend, silica, and NR/XSBR/Si composites.

Morphological Properties of NR/XSBR/Si Composites

The morphological properties of silica, NR/XSBR, and NR/XSBR blended with silica at different contents are shown in Figure 4. The surface fracture obtained from tensile testing of NR/XSBR showed a slightly rough surface when compared with NR/XSBR blended with silica at different contents. All NR/XSBR blended with silica showed similar rough surfaces. This led to the slight difference in elongation at break of NR/XSBR and NR/XSBR blended with silica at different contents since fracture surface can indicate failure manner which affected to the mechanical properties of samples. Silica particles were prepared by dropping acetic acid in CSS until neutral. This showed silica particles in a general form that is in spherical structure and partially agglomerated due to filler- filler interactions of silica (Choi, 2001; Stephen *et al*., 2007).

Thermal Properties of NR/XSBR/Si Composites

The thermal properties in term of degradation temperature of NR/ XSBR and NR/ XSBR blended with silica at different contents are listed in Table 1. The degradation temperature of NR/XSBR and NR/XSBR blended with silica at the different contents showed an insignificant difference in degradation temperature in the early weight loss levels of degradation. However, considering 70% weight loss level it was found that the degradation temperature of NR/XSBR/Si composites increased with the addition of silica, due to the higher thermal stability of silica than NR and XSBR.

Conclusions

The blending of NR and XSBR can improve the thermal properties of NR. NR/ XSBR at the ratio of 2:1 showed the highest tensile strength about 200% when compared with NR. The addition of silica into NR/XSBR at the ratio of 2:1 can improve the thermal properties of NR/XSBR blends with an insignificant difference in mechanical properties in terms of tensile strength and elongation at break. NR/XSBR at the ratio of 2:1 showed an increase in degradation temperature with increasing silica contents. Therefore, the best thermal properties were obtained from NR/XSBR/20Si composite.

Acknowledgments

The authors are grateful for financial supports from the Center of Excellence on Petrochemical and Materials Technology (PETROMAT) and chemicals for rubber compounding from the Chemical Innovation Co., Ltd.

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IUMRS-ICA 2021 IOP Publishing
Journal of Physics: Conference Series **2175** (2022) 012017 doi:10.1088/1742-6596/2175/1/012017

Mechanical and thermal properties of NR/XSBR composite reinforced with rice husk silica

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Abstract. Natural rubber (NR) is a renewable resource that is used in many products. In the production of NR products, other rubbers or fillers may be used to produce a product with required properties. However, most rubbers and fillers are synthetic which are non-environmentally friendly materials. To solve this problem, rice husk ash (RHA) from biomass power plant was used to prepare silica to be used as a filler in rubber by in-situ generation. The purer RHA was prepared by leaching with HCl to remove some metallics and increase silica contents by combustion. The purer RHA was dissolved in NaOH to obtain sodium silicate from RHA (RSS). Carboxylated styrene-butadiene rubber (XSBR) used as synthetic rubber was blended with NR in latex form. NR/XSBR at the ratio of 2:1 was mixed with RSS to obtain NR/XSBR/RSS mixture. Acetic acid was dropped into the mixture until neutral for precipitating silica to obtain NR/XSBR/RSi composite. The mechanical, morphological, and thermal properties of NR/XSBR/RSi composites at different contents of silica (5, 10, and 15 phr) were studied. The NR/XSBR/RSi composite with optimum content was compared with NR/XSBR/CSi composite which prepared silica from commercial sodium silicate (CSS) on mechanical, morphological, and thermal properties.

Keywords: Natural rubber latex, Carboxylated styrene-butadiene rubber latex, Rice husk ash, Silica, Rubber composite

1. Introduction

Natural rubber (NR) is one of Thailand's important renewable resource which can be used to produce in many products because it has high resilience, good tensile strength, tear resistance, etc. (1, 2). This is the reason of NR products covering in many industries such as automotive, medical, construction, etc. (3).

However, it is known that NR has low resistance to sunlight, ozone, and oxygen or heat aging because it consists of double bonds in the molecular structure (4). Therefore, the production of most NR products is not used only NR but also used it with other rubbers or fillers to obtain the required product properties (5, 6). For example, the tire manufacturers used NR with butadiene rubber (BR) or

styrene-butadiene rubber (SBR), carbon black (CB), and silica to improve wear, traction, rolling resistance, etc. (7, 8).

In the other rubbers and fillers, carboxylated SBR (XSBR) and silica are interesting materials because they can improve some NR properties such as mechanical and thermal properties. There are several reports mentioned that NR blended with XSBR which can easily modify the mechanical properties of NR and the incorporation of XSBR into NR can improve the thermal properties and gas permeability of NR (9). Poompradub et al. reported that filling NR with in-situ silica can improve not only the mechanical properties but also the thermal properties and it is superior compared with using ex-situ silica (10). However, it is known that these are synthetic that are non-environmentally friendly materials. Therefore, this research is to study in reducing this problem by using rice husk ash (RHA) from biomass power plant to prepare silica used as a filler in rubber by in-situ generation.

RHA is one of agricultural wastes that consists of high silica content in amorphous form that can be used in many applications such as adsorbent, catalyst, fertilizer, etc. (11). In-situ silica has several advantages, including enhanced silica dispersion, higher silica loading in the matrix, possible to customize the particle size, and energy-efficient processing (12).

This research aims to study the effect of rice husk silica (RSi) contents in NR/XSBR blend on mechanical, morphological, and thermal properties. The effect of silica sources was also studied by comparison between NR/XSBR/RSi composite with NR/XSBR/CSi composite in which silica prepared from commercial sodium silicate (CSS) at the same silica content on mechanical, morphological, and thermal properties.

2. Materials and Methods

2.1. Materials

Natural rubber latex (NRL) with 60wt% dry rubber content (DRC) and high ammonia (HA) treated with latex was purchased from Chemical & Material Co., Ltd. Carboxylated styrene-butadiene latex (XSBRL) was purchased from Jorakay Corporation Co., Ltd. Rice husk ash (RHA) from biomass power plant was purchased from Chia Meng Co., Ltd. Commercial sodium silicate (CSS) was purchased from Sigma-Aldrich Co., Ltd. Hydrochloric acid (HCl) was purchased from RCI Labscan Co., Ltd. Sodium hydroxide (NaOH) and acetic acid (CH_3COOH) were purchased from Carlo Erba Reagents. Stearic acid (SA), zinc oxide (ZnO), N-Cyclohexyl-2-Benzothiazole Sulfonamide (CBS), and sulfur (S) were supported by Chemical Innovation Co., Ltd.

2.2. Methods

2.2.1. Preparation of Sodium Silicate from RHA (RSS). RHA was purified by leaching RHA from biomass power plant with 1M HCl at 90°C by using magnetic stirrer for 3 h to remove some metallics. In the end of this process, the filter paper was used to filter unreacted RHA and DI water was used to wash unreacted RHA several times until the pH of unreacted RHA is neutral. And then, unreacted RHA was dried by using hot air oven at 110°C for 12 h and combusted by using muffle furnace at 600°C for 6 h to increase silica contents. RSS was prepared by dissolving the purified RHA that obtained from previous process with 1M NaOH at 90°C by using magnetic stirrer for 3 h and filter paper was used to filter unreacted RHA until obtain the clear solution that is RSS.

2.2.2. Preparation of NR/XSBR and NR/XSBR/Si Sheets. NR/XSBR/RSi sheets were prepared by mixing NRL to XSBRL at the ratio of 2:1, and RSS at various contents of silica (5, 10, and 15 phr) by using magnetic stirrer for 30 mins to obtain NR/XSBR/RSS mixtures. Acetic acid was dropped into NR/XSBR/RSS mixture until the pH of NR/XSBR/RSS mixture is neutral to precipitate silica. And then, the previous mixture was poured onto Teflon tray and dried at 60°C by using hot air oven for 72 h. For NR/XSBR/CSi sheet was prepared by using CSS at optimum silica content instead of RSS and following this process for comparing properties of NR/XSBR/Si composite at the same silica content

whereas silica was obtained from different sources. NR/XSBR sheet was also prepared by following this process whereas it was without sodium silicate mixing.

2.2.3. Preparation of NR/XSBR Blend and NR/XSBR/Si Composites. NR/XSBR and NR/XSBR/Si compounds were prepared by mixing NR/XSBR and NR/XSBR/Si sheets with 2 phr SA, 5 phr ZnO, 2.5 phr CBS, and 1 phr S by using two roll rubber mill. And then, the previous compound was vulcanized by using compression molding at 150°C with optimum curing time (t_c90) of each compound that was determined by Moving Die Rheometer (MDR, Gotech M2000) to obtain NR/XSBR blend and NR/XSBR/Si composites.

2.2.4. Chemical Properties. Energy Dispersive X-ray Fluorescence (EDXRF, Horiba XGT-5200) was used to characterize the chemical compositions of RHA, RSi, and CSi. RSi and CSi were prepared by dropping acetic acid into RSS and CSS was under magnetic stirrer until the pH of solution to neutral that grained silica precipitating. And then, the previous substance was dried at 110°C by using hot air oven for 24 h to obtain silica from RSS and CSS.

2.2.5. Mechanical Properties. The tensile properties of NR/XSBR blend and NR/XSBR/Si composites were measured according to ASTM D412 by using Universal Testing Machine (UTM, Instron 5565) with load cell of 5 kN and crosshead speed of 500 mm/min on the standard dumbbell specimen. At least five specimens were tested to obtain the average value for tensile strength, percentage elongation, modulus at 100% (M100), and 300% elongation (M300). The hardness of NR/XSBR blend and NR/XSBR/Si composites were measured according to ASTM D2240 by using hardness tester (Bareiss, HPE II) with test method that is Shore A. It makes five determinations of hardness at different positions on the specimen at least 6 mm in thickness to obtain the average hardness value.

2.2.6. Morphological Properties. The tensile fracture surface of NR/XSBR blend, NR/XSBR/Si composites, and silica particles that precipitated from RSS and CSS were coated with gold before examined by Field Emission Scanning Electron Microscope (FE-SEM, Carl Zeiss Auriga).

2.2.7. Thermal Properties. Thermogravimetric analyzer (TGA, Mettler Toledo TGA/DSC 1) was used to analyze the thermal decomposition of NR/XSBR blend and NR/XSBR/Si composites that placed in alumina pan and heated from room temperature up to 800°C under nitrogen with heating rate of 10°C/min.

3. Results and Discussion

3.1. Chemical Properties of Silica

The chemical compositions of RHA, RSi, and CSi are listed in Table 1. RHA from biomass power plant shows the lowest percentage SiO_2 composition is 82.97%. RSi shows percentage chemical compositions are similar to CSi and contain percentage SiO_2 composition approximately 97% indicating the RHA that can preparate silica similarity to commercial grade.

Table 1. Chemical compositions of RHA, RSi, and CSi.

	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	MnO_2	Fe_2O_3
RHA	-	82.97	9.78	3.31	0.13	2.68	1.13
RSi	2.27	97.20	0.34	0.10	0.03	-	0.05
CSi	2.38	97.28	0.11	0.06	0.06	-	0.11

3.2. Morphological Properties of Silica

The morphological images of RSi and CSi are shown in Figure 1. RSi and CSi show silica particles with a size that is lower than 100 nm in a general form that is spherical structure and partially agglomerate owing to filler-filler interactions of silica (13, 14). On the other hand, RSi shows smaller particle size and lower agglomeration than CSi.

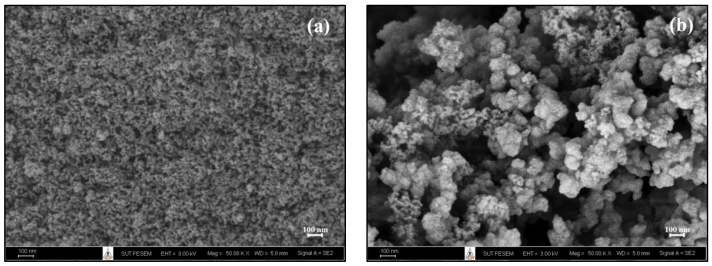
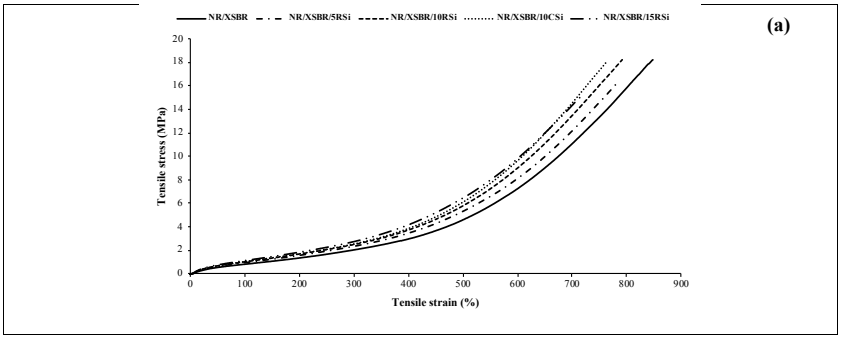


Figure 1. SEM images of (a) RSi and (b) CSi.

3.3. Mechanical Properties of NR/XSBR/Si Composites

The mechanical properties in terms of M100, M300, tensile strength, percentage elongation, and hardness of NR/XSBR blend and NR/XSBR/Si composites that blended with RSi at 5, 10, and 15 phr and CSi at 10 phr are shown in Figure 2. NR/XSBR blend shows the highest percentage elongation approximately 840% indicating the general rubber properties that is an elastic characteristic. The containing silica at 10 phr in NR/XSBR blend can improve the tensile strength whereas silica sources show insignificant effect on this property. Nevertheless, NR/XSBR/Si composites show the percentage elongation slightly decrease. The increasing silica contents in NR/XSBR blend show the mechanical properties in terms of M100, M300, and hardness are higher values than NR/XSBR blend because fillers with a higher stiffness than the matrix can increase the modulus of composite and value is higher when fillers increasing (15) whereas the silica sources also show insignificant effect on these properties.



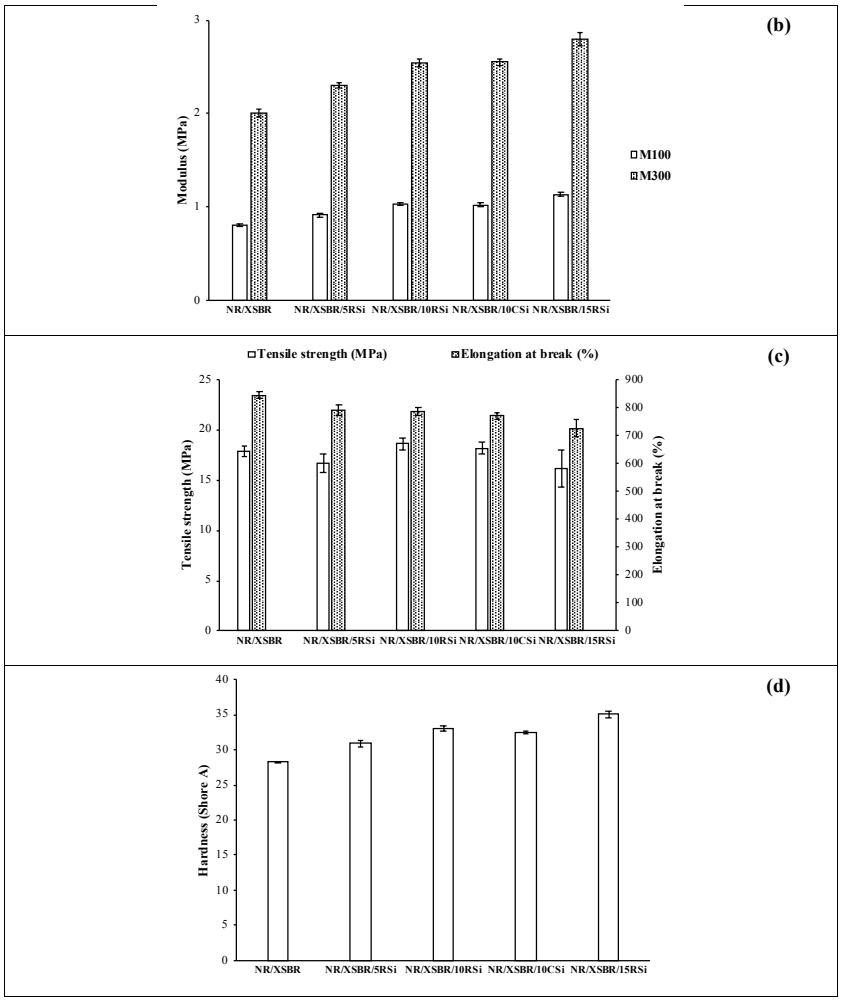


Figure 2. Mechanical properties of NR/XSBR blend and NR/XSBR/Si compositions showing (a) stress-strain curve, (b) modulus at 100% elongation and modulus at 300% elongation, (c) tensile strength and percentage elongation, and (d) hardness.

3.4. Morphological Properties of NR/XSBR/Si Composites

The morphological images of NR/XSBR blend and NR/XSBR/Si composites that blended with RSi at 5, 10, and 15 phr and CSi at 10 phr are shown in Figure 3. NR/XSBR blend shows holeless on tensile

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Journal of Physics: Conference Series **2175** (2022) 012017 doi:10.1088/1742-6596/2175/1/012017

fracture surface caused by filler detachment due to incompatibility between silica and rubber. In the same way, NR/XSBR/15RSi composite shows the highest holes number with a large-sized on tensile fracture surface owing to filler agglomeration that relates the lowest percentage elongation. The tensile fracture surfaces of NR/XSBR/10RSi and NR/XSBR/10CSi composites shows analogous images.

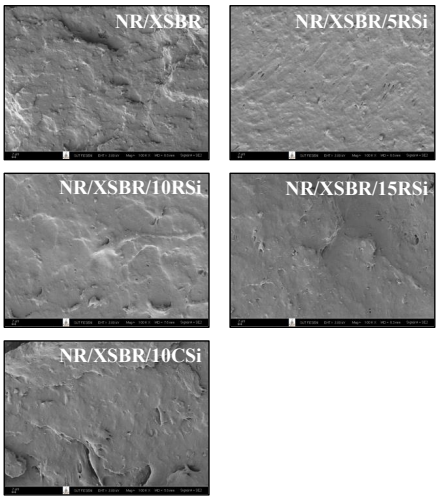

Figure 3. SEM images of NR/XSBR blend and NR/XSBR/Si composites.

3.5. Thermal Properties of NR/XSBR/Si Composites

The thermal properties in terms of degradation temperature at onset (T_{onset}), temperature at 50% weight loss level ($T_{50\%}$), temperature at maximum weight loss level (T_{max}), percentage residue, and percentage weight loss level at maximum degradation temperature (%wt. loss at T_{max}) of NR/XSBR blend and NR/XSBR/Si composites blended with RSi at 5 and 10 phr and CSi at 10 phr are listed in Table 2. The containing silica in NR/XSBR blend can improve the thermal stability of rubber as shown clearly in terms of $T_{50\%}$, T_{max}, and %wt. loss at T_{max}. $T_{50\%}$ and T_{max} of NR/XSBR/Si composites are higher temperatures than NR/XSBR blend whereas %wt. loss at T_{max} of NR/XSBR/Si composites lower than that of NR/XSBR blend. This may be due to the strong interaction of silanol group of silica with carboxylic group of XSBR. On the other hand, the contents and sources of silica play insignificant effect on the degradation temperature of NR/XSBR/Si composites.

Table 2. Thermal characteristics of NR/XSBR blend and NR/XSBR/Si composites.

Sample	T_{onset} (°C)	$T_{50\%}$ (°C)	T_{max} (°C)	%wt. loss at T_{max} (%)	Residue (%)
NR/XSBR	351	390	387	47.36	4.13
NR/XSBR/5RSi	352	400	392	43.75	9.40
NR/XSBR/10RSi	353	403	393	42.44	12.00
NR/XSBR/10CSi	352	400	391	43.49	14.67

4. Conclusions

Silica can be successfully prepared from RHA which comparable to commercial grade containing 97% SiO_2 composition and silica particle size is lower than 100 nm. The increasing silica contents in NR/XSBR blend improve modulus and hardness. NR/XSBR/10Si composites show the highest tensile strength. Silica sources show insignificant effect on mechanical properties. Higher decomposition temperature of NR/XSBR was obtained with the addition of silica.

Acknowledgments

The authors are grateful for financial support from the Center of Excellence on Petrochemical and Materials Technology (PETROMAT) and chemicals support that used in rubber compounding from the Chemical Innovation Co., Ltd.

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